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Mechanism of grain growth in phosphorus-doped polycrystalline silicon thin films during annealing

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Abstract : In the present investigation, a kinetic model has been proposed based on the thermodynamical concepts to determine the surface area of the grain in heavily phosphorus doped polysilicon thin film for various dopant concentration, annealing time and temperatue of annealing The numerical analysis for our theoretical predictions has been made using computer simulation technique. The simulated values are compared with available experimental reports. This model has been extended to determine the grain size distribution for various dopant concentration, annealing time and temperature The results are discussed in detail.

Keywords : Grain growth, phosphorus doped polysilicon, kinetic model

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1. Introduction

Grain boundary migration is the movement of a grain boundary perpendicular to the tangent plane. Therefore, boundary migration is based, in principle, always on the movement of atoms from one grain to the other across the grain boundary [1]. All the reported theories [2–5] for grain boundary migration assigned that the elementary process of grain boundary migration is the diffusion of one atom or a group of atoms from the surface of the shrinking grain to the surface of the growing grain. The different atomic jumps were considered to be independent of each other. Therefore, the calculated migration rate is independent of the boundary orientation relative to the grains. Lucke and coworkers [6,7] have reported the 0.01% of manganese or iron can decrease the rate of recrystallization of high purity aluminium by factors 10^{12} or 10^{16} respectively.

Polycrystalline silicon layers are extensively used in the fabrication of high value resistors, gate electrodes and diffusion source for both active devices as well as contacts. Thermal treatment has been applied for the fabrication of devices. Many experimental results [8–14] on grain growth in polycrystalline silicon are available. But concerning the grain growth mechanism, only a few results [15–21] have been reported. Therefore, understanding the role of impurities is absolutely essential for the understanding of grain growth mechanism in polycrystalline silicon. In the present communication, we have proposed a kinetic model to determine the grain surface area in heavily phosphorus doped polycrystalline silicon for different phosphorus concentration, annealing time and temperature.

Many theoretical reports on grain size distribution are available [22–29], but there are no reports on the grain size distribution in heavily phosphorus doped polysilicon during high temperature annealing except the work [18]. In the second part of this paper, the model has been extended to determine the grain size distribution in heavily phosphorus doped polysilicon for different dopant concentration, annealing time and annealing temperature based on the thermodynamical concepts. The results are discussed in detail.

2. Theory

The driving force leading to grain boundary migration in polycrystalline materials is determined not only by the physical constants characteristic of the substances in bulk, but also by the shapes and dimensions of the individual grains. The driving force for the movement of atoms from one grain to another [17] is

$$E = \frac{Sa^3G}{r}, \tag{1}$$

where S is a constant related to the geometric shape of the grains (for spherical grains $S = 2$), G is the grain boundary energy, a is the lattice constant and r is the grain size.

From basic rate theory, the net rate of atomic transfer across the boundary (from lattice sites of one grain to those of a neighbour) is given by [11]

$$l = D_g/J^2\left[1 - \exp(-E/kT)\right], \tag{2}$$

where D_g is the self diffusion constant of the atoms across the grain boundary, k is the Boltzmann's constant, T is the annealing temperature and J is the thickness of the grain boundary. The rate of boundary motion is given by the product of the net rate of atomic transfer across the boundary and the thickness. Therefore, the grain growth rate becomes

$$dr/dt = lJ = D_g/J\left[1 - \exp(-E/kT)\right]. \tag{3}$$

Expanding the exponential, neglecting the cube and higher powers of E/kT and using eq. (1)

$$dr/dt = 2D_g/J\,(L/r)\left[1 - (L/r)\right], \tag{4}$$

where $L = Sa^3G/2kT$

Integrating eq. (4)

$$2D_g Lt/J = \left[(r-L)^2/2 + 2L(r-L) + L^2 \log(r-L)\right]_{r_0}^{r}, \tag{5}$$

where t is the time of annealing and r_0 is the initial grain size. Expanding the right hand side of eq. (5), applying the limits and neglecting the cube and higher power of L, since L is very small compared to the value of r_0, we get

$$r = (r_0 + L)\left[\left(1 + 4D_g Lt/J(r_0 + L)^2\right)^{1/2} - L/(r_0 + L)\right]. \tag{6}$$

It is noted that the grain size is directly proportional to the square root of annealing time as reported earlier [9,11,17–20]. Surface area of the grain can be written from eq. (6) as

$$A = A_0\left[1 + 4D_g Lt/Jr_0(L + r_0)\right], \tag{7}$$

where A_0 is the initial grain surface area. It is apparent from eq. (7), that the grain surface area increase linearly with the annealing time. The rate at which the grain surface area increases with time is the strong fraction of annealing temperature and dopant concentration through the relation

$$\text{Slope} = 4A_0 D_g L/Jr_0(L + r_0).$$

This model has been extended to evaluate the grain size distribution for different annealing time and annealing temperatures. By introducing a dimensionless variable known as relative grain size, $R = r/r_{cr}$ in eq. (4) and simplifying

$$dR^2/d\tau = \left[\left(4D_g L/J\right)(1 - L/Rr_{cr})dt/dr_{cr} - R^2\right], \tag{8}$$

where $1/d\tau = r^2/dr_{cr}$, r_{cr} is the average grain size and $d\tau$ represents the time.

$$dR^2/d\tau = \left[X(1 - Y/R) - R^2\right],$$

where $X = \left(4D_g L/J\right) dt/dr_{cr}$ and $Y = L/r_{cr}$

$$dR/d\tau = 1/2R\left[X(1 - Y/R) - R^2\right] \tag{9}$$

The eq. (9) has been solved to determine the steady state grain size distribution during normal grain growth mechanism, using the Hillert's [23] approach. The whole distribution of the individual grain size during the steady state is given by

$$P(R) = 1/2 \exp(-\beta\psi/2)/[dR/d\tau], \tag{10}$$

where β is a constant ($\beta = 2$ for two dimensional system $\beta = 3$ for three dimensional system) and

$$\psi = \int^{R} dR/(-dR/d\tau) \tag{11}$$

From eqs. (9) and (11)

$$\psi = \left[2/3 \log(R^3 - XR + XY)\right]_0^R + W, \tag{12}$$

where $$W = 2X/3 \int_0^R dR/(R^3 - XR + XY).$$

Numerical integration method has been followed to determine the value of W. Substituting the values of $dR/d\tau$ and ψ from eqs. (9) and (12) respectively in eq. (10), we get

$$P(R) = \beta R^2/(R^3 - XR + XY)\left[(R^3 - XR + XY)/XY\right]^{\beta/3} \exp(\beta W/2). \tag{13}$$

Eq. (13) gives the grain size distribution.

3. Computer programme

Computer simulation technique has been used to evaluate the numerical values of our theoretical findings. The grain boundary self diffusion of silicon atoms is function of impurity concentration in the grain and the grain boundary free energy changes with the grain size. To determine the grain boundary self diffusion of silicon atoms, we have followed the method used by Mei *et al* [9] to our expression for the grain size eq. (6) and grain surface area given by eq. (7). The grain boundary self diffusion has been determined for each time step of one minute from the size of previous time step for the given doping concentration and annealing temperature. Using the grain boundary self diffusion values, the size of the grain at a given time step is evaluated. This process is reiterated until the time of annealing is achieved. The size distribution of grain in the polysilicon at the end of each time is also evaluated using eq. (13).

4. Results and discussion

The simulated numerical values of the radius of the grain and surface area of the grain in polysilicon doped with phosphorus have been presented in Figures (1–6). Figure 1 depicts the radius and surface area of the grain *vs* annealing temperature for four different dopant concentrations and for one hour of annealing time. It is observed that the radius and surface area of the grain increases with dopant concentration and annealing temperature. The reported experimental values [9] for the radius of the grain have also been plotted in the graph. The activation energy for the grain growth is found to be 0.5028 eV. Figure 2 is drawn for the radius and surface area of the grain as a function of annealing temperature for different annealing times and for phosphorus concentration of 6×10^{25} atoms/m^3. It is observed from the graph that the grain increases with annealing time for all the annealing temperatures. Figure 3 shows variation of the radius and surface area of the grain with the dopant concentration for various annealing time and for the given annealing temperature of

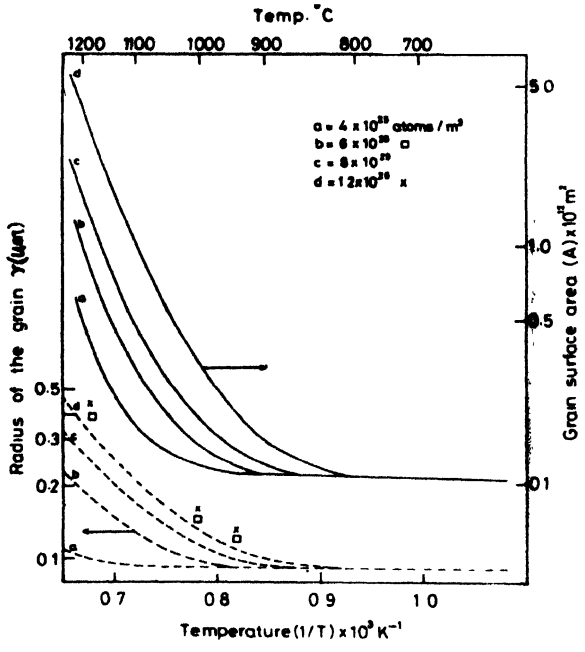

Figure 1. Radius of grain (– – –) and surface area of the grain (——) *vs* annealing temperature of *P* doped polysilicon after one hour annealing for various values of dopant concentration. Available experimental reports [8] are given (*X*, □).

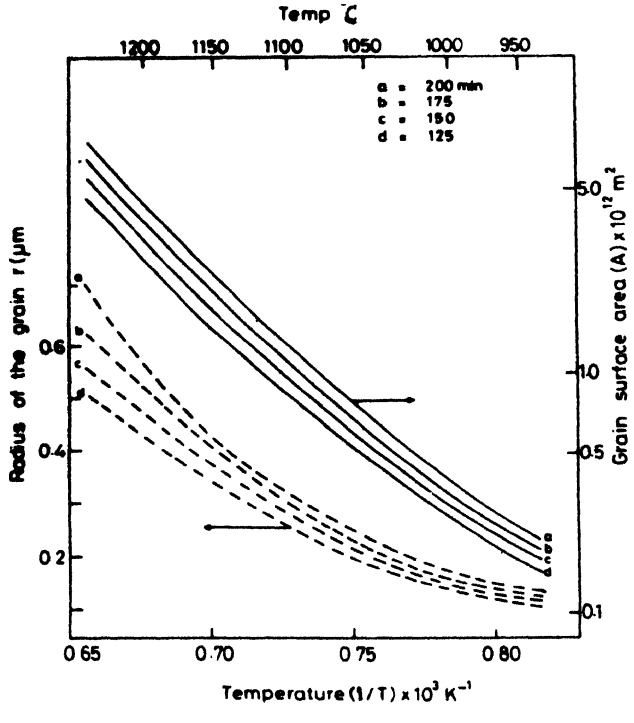

Figure 2. Variation of radius and surface area as a function of annealing temperature for various annealing times and for *P* concentration of 6×10^{25} atoms/m^3.

1173 K. Figure 4 depicts the radius and surface area of the grain *vs* dopant concentration for different annealing temperature. It reveals that the grain size and surface area of the grain

increases with annealing temperature. The reported experimental values [9] for radius of the grain have also been plotted in the graph. Figure 5 shows the radius of the grain and surface

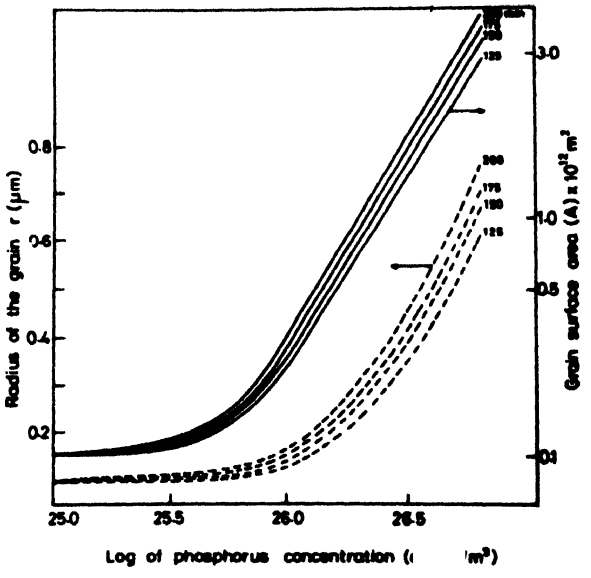

Figure 3. Radius of the grain and surface area with function of concentration for different annealing times and for annealing temperature of 1173 K.

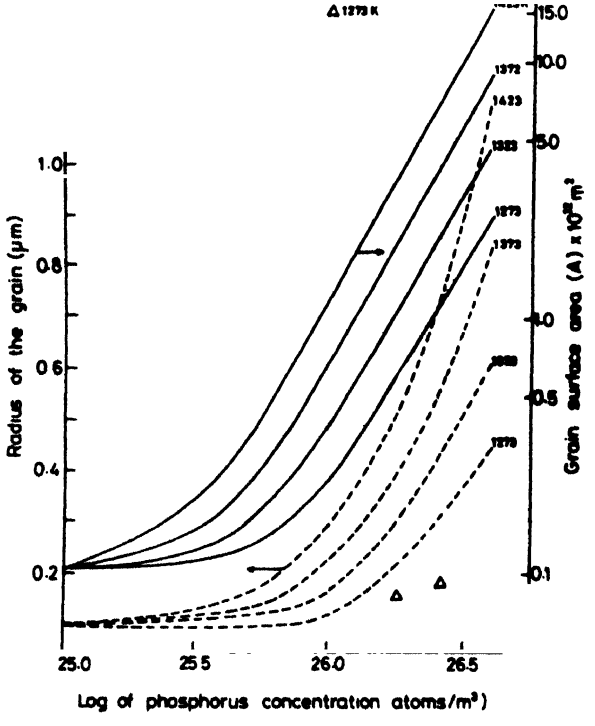

Figure 4. Function of radius of the grain and surface area with concentration after one hour of annealing for different annealing temperatures. Available experimental reports [8] are given (Δ).

area *vs* annealing time for various annealing temperature and for the phosphorus concentration of 6×10^{25} atoms/m^3. Figure 6 depicts the radius of the grain and its surface

area *vs* annealing time for different phosphorus concentration and for one hour of annealing and annealing temperature of 1273 K.

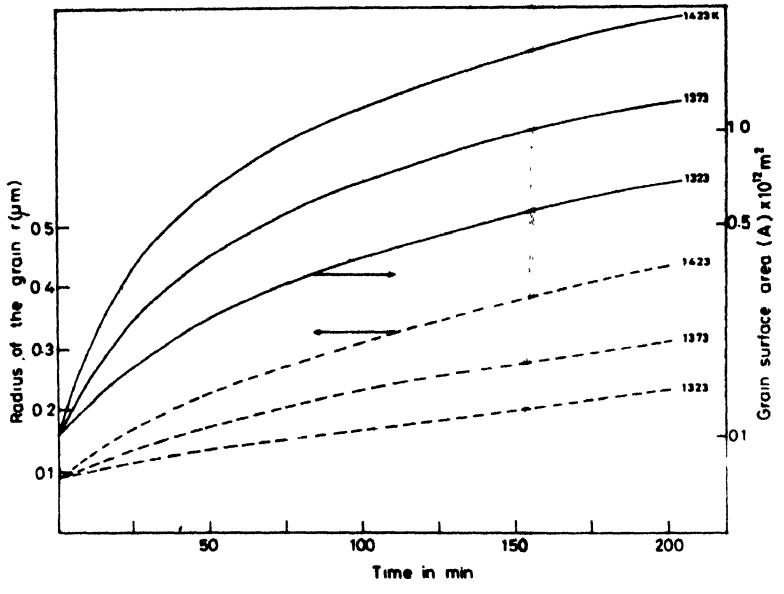

Figure 5. Variation of radius of the grain and surface area with annaling time for different annealing temperatures and for P concentration of 6×10^{25} atoms/m^3.

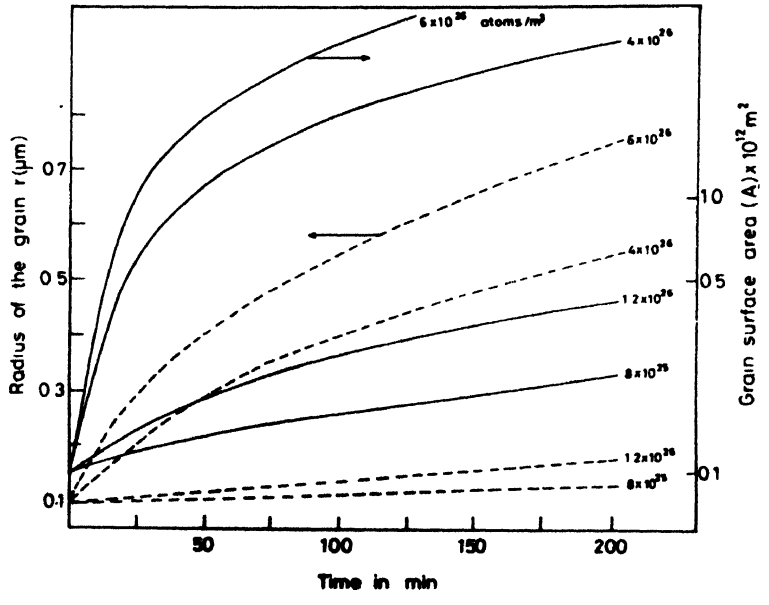

Figure 6. Radius of the grain and surface area *vs* annealing time for different P concentrations and annealing temperature 1273 K.

Figure 7 shows the grain size distribution *vs* relative grain size for the annealing temperature of 1273 K, for the phosphorus concentration of 8×10^{25} atoms/m^3 and

for three different annealing times. From the graph it is concluded that the grain size distribution increases with annealing time. Dotted line in the Figure 7 shows the

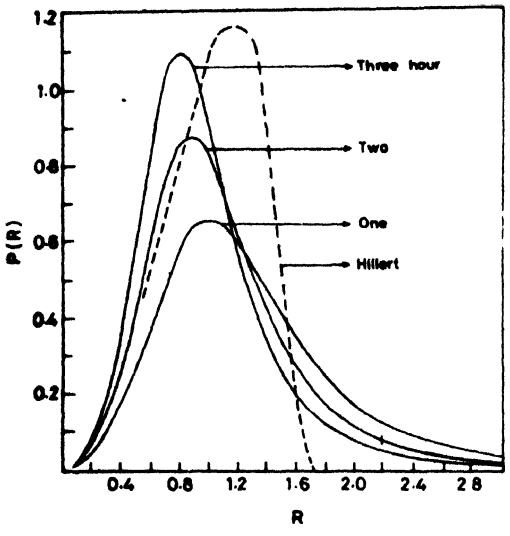

Figure 7. Variation of grain size distribution *vs* relative grain size for annealing temperature of 1273 K and *P* concentration 8 × 10^{25} atoms/m^3, for three different annealing times. Dotted line shows Hillert's report.

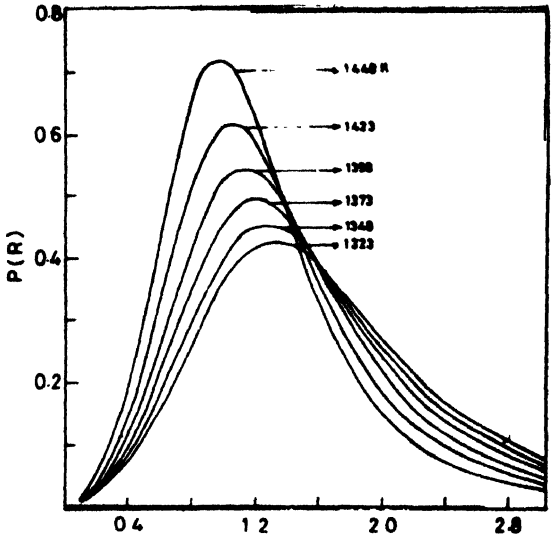

Figure 8. Grain size distribution *vs* relative grain size for one hour of annealing and *P* concentration of 6 × 10^{25} atoms/m^3 for six different annealing temperatures.

Hillert's [23] result. Figure 8 depicts grain size distribution *vs* relative grain size for one hour of annealing and phosphorus concentration of 6 × 10^{25} atoms/m^3 and for six different

annealing temperatures. It is observed that the grain size distribution increases with annealing temperature. Figure 9 is plotted between the grain size distribution and relative

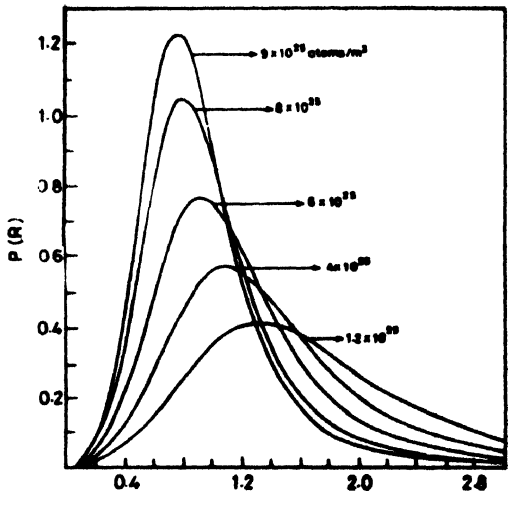

Figure 9. Grain size distribution *vs* relative grain size for two hours of annealing and annealing temperature 1273 K for five different P concentrations.

grain size for two hours of annealing and annealing temperature of 1273 K for five different phosphorus concentrations. From the Figures (7–9) it is observed that the grain size distribution slowly increases with increase of relative grain size, and attains a maximum, then with further increase of relative grain size, the grain size distribution decreases. The grain size distribution increases with phosphorus concentration, annealing time and annealing temperature. The reason attributed is due to the large amount of phosphorus atoms segregated at the grain boundaries.

5. Conclusions

We have concluded that the radius, surface area and size distribution of the grain in polysilicon doped with phosphorus increases with dopant concentration, annealing time and annealing temperature.

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